Exchange constant variation effects in magnetocaloric and barocaloric isothermal potentials

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Frequently, magnetothermal studies of many ferromagnetic materials are focused considering the system such as a paramagnet subject to a molecular field being characterized by an exchange constant. However, magnetostructural transitions can produce abrupt changes in the exchange constant triggering magnetic transitions of first order at transformation temperatures, which can be shifted by applying magnetic field or pressure. On thermodynamic bases we show how an abrupt but continuous variation of the exchange constant at the transition can play an important role in the magnetocaloric and barocaloric isothermal potentials in systems showing structural transformation.

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INTRODUCTION

The magnetocaloric effect (MCE), i.e., the change in temperature of a magnetic material resulting from a change in applied magnetic field, is the basis for magnetic refrigeration mainly as an emerging technology for room-temperature applications. This effect can be characterized by means of the isothermal magnetocaloric potential, $\Delta S^M(T)$, which can be calculated from magnetic data. In this case one can use the derivatives $(\partial M/\partial T)_{H,P}$, for a magnetic field range and fixed temperatures, to obtain $\Delta S^M(T)$ using the Maxwell's differential equation $(\partial S/\partial H)_{T,P}=(\partial M/\partial T)_{H,P}$. Large values of these derivatives, as that expected in the case of first-order magnetic transitions, at particular temperatures and for lower fields (H less than approximately 2 T) implicates the possibility to obtain very high $\Delta S^M(T)$ intensities. In this frame it is of great importance the understanding of the mechanisms involved in the establishment of these conditions, mainly to the search for new MCE materials.

Since the discovery of the giant magnetocaloric effect (GMCE) at room temperature in $Gd_5Si_2Ge_2$ compound,[1] considerable efforts are being made to improve the effect in this compound and also in new MCE materials. Numerous doping or partial substitution experiments with nonmagnetic elements, as well as subjected to pressure, has been revealing interesting results. For instance, in $Gd_5Si_{1.95-x}Ge_{2.05-x}Sn_{2x}$ ($2x \leqslant 0.10$) compounds the first order magnetic and structural transition, also observed in $Gd_5Si_2Ge_2$, is retained. In particular, for $2x=0.03$, an increase in both T_C (from 265 to 277 K) and MCE peak (from 15 to 28.9 J/kg K for a magnetic field change of 2 T), in respect to the parent compound, have been reported.[2] For partial substitution, in $Gd_5Ge_2(Si_{1-x}Sn_x)_2$ compound with $x=0.40$, the first order transition temperature is shifted to low temperatures ($\sim$180 K) and the $\Delta S^M(T)$ reaches 33 J/kg K for a magnetic field change of 5 T.[3] In the $Gd_5Si_2Ge_2$ compound under pressure, the position of the ΔS^M peak is shifted to high temperatures and its intensity is decreased. Additionally, a change is observed from first to second order in the nature of the transition for pressures of about 6 kbar.[4] In the same way the MnAs compound has been extensively studied[5–7] and also other important materials have received much interest.[8,9]

In all cases, in principle, $\Delta S^M(T)$ is built with a direct or indirect role of the lattice and electronic structure over the magnetization, and those can contribute also to the total entropy change (see, for instance, Refs. 10 and 11). Advanced MCE materials[12] presents giant magnetostriction and it would be correlated with their intense MCE still observed for small magnetic field changes ($\Delta H \sim 2$ T). However, the role of the volume magnetostriction in the MCE is not completely understood and further complicated when magnetostructural transition (MST) is realized.

Some attempts were performed in order to include the effect of magnetostructural transition in the magnetic behavior of the MnAs compound.[13,14] Recently, a study to understand the origin of MST in the $Gd_5Si_2Ge_2$ compound has been reported.[15] In this case, the transition is related to a change in bonds that can be reversibly induced by changing the external parameters but with a noticeable hysteresis. The entropy change of the structural transformation, $\Delta S^{st}(T)$, would be the "lattice part" of the total isothermal one. More recently,[12] $\Delta S^{st}(T)$ appears maintaining its original interpretation,[16] i.e., it has been recognized as a part of the isothermal magnetic entropy change.

From thermodynamics in a mean field approximation, considering volume-dependent energy terms together with magnetic terms, an important lattice (phonon) contribution to the total entropy change in a system presenting MST was proposed.[17,18] In another interesting approach,[19] a nonmean field model uses a Hamiltonian where the usual elastic term supports the lattice structure whereas an original term supports a more regular lattice. In a specific choice of the variation of the exchange interaction, one can suppose an abrupt change on its strength from weak to strong intensity and vice versa. The result is a first order transition characterized by a sharp variation in volume and magnetization of the system.

In another work[20] we proposed that the entropy related only to magnetism can overcome the conventional magnetic limit, $R\ln(2J+1)$, R being the molecular gas constant and J the total angular momentum of the magnetic ions. As a consequence, the isothermal magnetic entropy change, $\Delta S^M(T)$, could reach intense values given origin to giant and colossal magnetocaloric effect (CMCE), which were observed experimentally in a number of compounds.[1,5,21,22,27] We stressed that there is an additional-exchange entropy contribution that makes it possible to obtain high values of the $\Delta S^M(T)$ peak, which comes from a field-induced abrupt change of the

exchange-constant related to MST at each fixed temperature. It is important to notice that the variation of the exchange-constant has been considered in some magnetostriction studies.[23,24]

Using thermodynamics, we show below how the variation of exchange constant is important to account for an intense MCE. In addition, we extend our analysis introducing pressure to obtain the additional-exchange contribution to the barocaloric effect (BCE). For this case, one can use the derivatives $(\partial V/\partial T)_{H,P}$, for a pressure field range and fixed temperatures, to calculate $\Delta S^V(T)$ using the Maxwell's differential equation $(\partial S/\partial P)_{H,T}=-(\partial V/\partial T)_{H,P}$.

THEORY

Fixed pressure

In his Nobel Lecture, Louis Néel[25] emphasized, for the energy molecular field H_m, the relation $H_m=h-T\partial h/\partial T$, with $h=\eta M$ the well-known Weiss molecular field[26] where η is the exchange constant and M is the magnetization. Also, through the internal energy, $dU=-H_m dM$, and using the second law of thermodynamics, the elemental entropy results $(\partial h/\partial T)dM$. One can extend this result by considering the entropy as the function $S(M,h)$. Then, the elemental exchange-entropy can be written as

$$dS^e=\left(\frac{\partial S^e}{\partial M}\right)_h dM+M\left(\frac{\partial S^e}{\partial h}\right)_M d\eta. \tag{1}$$

The first term on the right-hand side of Eq. (1) is the above mentioned elemental entropy, easily recognized from the Maxwell's relation applied to exchange, $(\partial S^e/\partial M)_h=-(\partial h/\partial T)_M$. For the second one the alternative Maxwell's relation $(\partial S^e/\partial h)_M=(\partial M/\partial T)_h$ can be used. The superscript "e" denotes exchange.

The dependences $\eta(H,T,P)$ and $M(H,T,P)$ leaves from (1), for T and P fixed, to

$$dS^e=M\left(\frac{\partial\eta}{\partial T}\frac{\partial M}{\partial H}-\frac{\partial\eta}{\partial H}\frac{\partial M}{\partial T}\right)dH \tag{2}$$

with $S^e(H,T,P)$ in the above situation one can obtain

$$\left(\frac{\partial S}{\partial H}\right)^e=\left(\frac{\partial M}{\partial T}\right)^e=M\left(\frac{\partial\eta}{\partial T}\frac{\partial M}{\partial H}-\frac{\partial\eta}{\partial H}\frac{\partial M}{\partial T}\right). \tag{3}$$

In the following, we use a formal description of magnetization to link $(\partial M/\partial T)^e$ with the total variation of magnetization with temperature.

It is well known that introducing an external field, H, the equation of state for ferromagnets becomes functionally defined, i.e., $M=f[(H+h)/T]$. Frequently, the Brillouin function $B_J(X)$ with argument $X=Y(H+\eta M)/T$ and $Y\equiv gJ\mu_B/k$, leads to a good description of magnetism through $M=YkB_J(X)$ with ηM constant, or equivalently, $\partial X/\partial T=-X/T$. In this case, the ferromagnet is seen as a paramagnet with the presence of a molecular field, and the magnetic entropy turns out to be given by $S^c=-k\int X\frac{\partial B_J(X)}{\partial X}dX$ with $dH=(T/Y)dX$. In the literature the equivalent expression $S^c=R[\ln Z+\langle E\rangle/kT]$ used to obtain the magnetic entropy change, $\Delta S^c=S^c(H_{\text{final}})-S^c(H_{\text{initial}})$, is currently found. Here Z is the partition function, $\langle E\rangle$ is the mean energy of the system, and the limit $R\ln(2J+1)$ at high temperatures is clearly established. The conventional character is denoted by the superscript "c".

In this way, considering ηM as a constant, one can write

$$\left(\frac{\partial S}{\partial H}\right)^c=\left(\frac{\partial M}{\partial T}\right)^c=\left(-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\right)^c, \tag{4}$$

which we termed as "*conventional*". This denomination suggests the inability to overcome the limit $R\ln(2J+1)$ imposed to the ΔS^M peak.

Now, taking into account the variation of ηM with external parameters, H and T, it is easy to show that

$$\frac{\partial M}{\partial T}=\frac{Y^3k^2\frac{\partial B_J(X)}{\partial X}\frac{B_J(X)}{T}\frac{\partial\eta}{\partial T}-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}}{1-Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}} \tag{5}$$

and using the relation (see Appendix A)

$$-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}=\left(-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\right)^c+Y^3k^2\left(\frac{\partial B_J(X)}{\partial X}\right)^2\frac{X}{T}\frac{\eta}{T}+Y^2k^2B_J(X)\frac{X}{T}\frac{\partial B_J(X)}{\partial X}\frac{\partial\eta}{\partial H}$$

one can obtain (see Appendix B) $\frac{\partial M}{\partial T}=\left(\frac{\partial M}{\partial T}\right)^c+\left(\frac{\partial M}{\partial T}\right)^e$, or equivalently, $\Delta S^M=\Delta S^c+\Delta S^e$. This result allows the possibility to overcome the conventional magnetic limit.

From magnetization data at fixed pressure, $M(H,T)$, the isothermal magnetocaloric potential, ΔS^M, for a field variation from 0 to H, can be determined through $\Delta S^M=\int_0^H(\partial M/\partial T)_{H'}dH'$. Applying this relation to experimental data, a colossal MCE has been obtained in both MnAs under hydrostatic pressure[27] and recently in $Mn_{1-x}Fe_xAs$ under ambient pressure.[22] The intense peak of ΔS^M observed at the transition can be attributed to the high values of $(\partial M/\partial T)_{H,P}$ established for low fields included in the interval of magnetic fields between 0 and H (usually 5 T).

Fixed magnetic field

Now, we are going to consider the role of the pressure. For this, one must take into account the implications of the change of the pressure on both the magnetization and the exchange constant. An important observation is that the dependence of the magnetization with pressure is conditioned to the dependence of the exchange constant with pressure. In fact, in a similar way to the deduction of Eq. (5), using the mean field model it is easy to show that

$$\frac{\partial M}{\partial P}=\frac{Y^3k^2\frac{\partial B_J(X)}{\partial X}\frac{B_J(X)}{T}\frac{\partial\eta}{\partial P}}{1-Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}}. \tag{6}$$

Evidently in a model that considers a crystalline rigid material (in which the exchange interaction is invariable) pres-

sure effects are null and Eq. (6) becomes relevant only when treated with elastic materials. If the interest is to study the equilibrium volume then the elastic energy must be considered explicitly.[13] We considered here that the equilibrium volume, $V(H,T,P)$, was established and the fundamental quantity is the corresponding value of the exchange constant. It is important to notice that Eq. (6) remembers the early one reported by Smoluchowsky in his theory of volume magnetostriction.[23]

The total entropy related to thermal expansion can be obtained from

$$\left(\frac{\partial S}{\partial P}\right)_T = -\left(\frac{\partial V}{\partial T}\right)_P$$

and includes phonon and electronic contributions in addition to magnetic contributions. In some materials the electronic contribution can become as important as in the case of the FeRh alloy.[28] In nonmagnetic materials showing structural transformations, one can wait for a non-negligible phonon contribution to the change of the total entropy. In relation to magnetic contributions to ΔS^V, the structural phase transitions induced by pressure are of interest mainly when magnetic parameters suffer great changes. To include the change of h, one can consider the volume as the function $V(M,h)$. In the same way as we obtained Eq. (2), in the present case we arrive to

$$dS^{V,e} = M\left(\frac{\partial \eta}{\partial T}\frac{\partial M}{\partial P} - \frac{\partial \eta}{\partial P}\frac{\partial M}{\partial T}\right)dP. \tag{7}$$

The exchange contribution to isothermal barocaloric potential, $\Delta S^{V,e}$, given by Eq. (7) and related to volume exchange striction, incorporates the cross derivatives of magnetization and exchange constant with respect to pressure and temperature. Then, like the additional-exchange contribution to the MCE, the exchange-constant derivatives can play an important role also to the BCE.

CONCLUSIONS

Extending the molecular field paradigm to include the $\eta(H,T,P)$ dependence, we showed that ΔS^M is the sum of two terms, ΔS^c and ΔS^e, the conventional and additional-exchange magnetic contributions, respectively. For the first contribution, in the literature the limit $R\ln(2J+1)$ for the $\Delta S^M(T)$ peak is recognized. The second one is related to the derivatives $\partial\eta/\partial T$ and $\partial\eta/\partial H$, which have to be evaluated for the isothermal points from H_{initial} to H_{final}, and makes it possible to overcome the conventional limit to the magnetic entropy change. The term $\partial\eta/\partial P$ does not appear in the calculus of the contribution to magnetocaloric isothermal potential but it evidently appears in the calculus of the barocaloric potential. For fixed pressure in the MCE or fixed field in the BCE, the exchange constant needs to be adjusted when the system crosses from one phase to another phase during the structural transformation. This is the central point of our theory which establishes that both caloric effects may benefit from a favorable change in the exchange energy. Details of the exchange-constant variation must be considered for each particular system subjected to structural transformation.

The relations proposed in this study based on the exchange-constant variation, for the magnetocaloric and barocaloric effects, can be applied in materials showing giant magnetostriction. This is the case of MnAs and some parent compounds which show magnetostructural transition at temperatures which can be shifted by field or pressure. The present results were applied to some of these compounds, e.g., $MnAs_{0.95}Sb_{0.05}$, and will be published elsewhere.[29]

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APPENDIX A

Taking into account the variation of ηM we have $dH = (T/Y)dX - \eta dM - M d\eta$. Then,

$$\begin{aligned}
-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}dH &= -Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\left(\frac{T}{Y}dX - \eta dM - M d\eta\right) \\
&= \left(-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\right)^c dH + Yk\frac{\partial B_J(X)}{\partial X} \\
&\quad \times \frac{X}{T}\left(\eta Yk\frac{\partial B_J(X)}{\partial X}\frac{\partial X}{\partial H} + YkB_J(X)\frac{\partial \eta}{\partial H}\right)dH \\
&= \left(-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\right)^c dH + Yk\frac{\partial B_J(X)}{\partial X} \\
&\quad \times \frac{X}{T}\left(\frac{\eta}{T}Y^2k\frac{\partial B_J(X)}{\partial X} + YkB_J(X)\frac{\partial \eta}{\partial H}\right)dH
\end{aligned}$$

for the last expression we used $\partial X/\partial H = Y/T$. Finally we obtain

$$\begin{aligned}
-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T} &= \left(-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\right)^c + Y^3k^2\left(\frac{\partial B_J(X)}{\partial X}\right)^2\frac{X}{T}\frac{\eta}{T} \\
&\quad + Y^2k^2B_J(X)\frac{X}{T}\frac{\partial B_J(X)}{\partial X}\frac{\partial \eta}{\partial H}.
\end{aligned}$$

APPENDIX B

From Eq. (5) and using the result of Appendix A we can write

$$\begin{aligned}
&\left(1 - Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}\right)\frac{\partial M}{\partial T} \\
&\quad = Y^3k^2\frac{\partial B_J(X)}{\partial X}\frac{B_J(X)}{T}\frac{\partial \eta}{\partial T} + \left(-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\right)^c \\
&\quad\quad + Y^3k^2\left(\frac{\partial B_J(X)}{\partial X}\right)^2\frac{X}{T}\frac{\eta}{T} + Y^2k^2B_J(X)\frac{X}{T}\frac{\partial B_J(X)}{\partial X}\frac{\partial \eta}{\partial H}.
\end{aligned} \tag{B1}$$

With $\frac{\partial M}{\partial T}=\left(\frac{\partial M}{\partial T}\right)^c+\left(\frac{\partial M}{\partial T}\right)^e$ and using the relations (3) and (4), the left part of Eq. (B1) becomes

$$\left(1-Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}\right)\frac{\partial M}{\partial T}=\left(1-Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}\right)\times\left[\left(-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\right)^c+\left(\frac{\partial M}{\partial T}\right)^e\right]. \tag{B2}$$

On the other hand, with the relation (5) and

$$\frac{\partial M}{\partial H}=\frac{Y^3k^2\frac{\partial B_J(X)}{\partial X}\frac{B_J(X)}{T}\frac{\partial \eta}{\partial H}+Y^2k\frac{\partial B_J(X)}{\partial X}\frac{1}{T}}{1-Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}}$$

in the proposed relation $\left(\frac{\partial M}{\partial T}\right)^e=M\left(\frac{\partial \eta}{\partial T}\frac{\partial M}{\partial H}-\frac{\partial \eta}{\partial H}\frac{\partial M}{\partial T}\right)$ we obtain

$$\left(\frac{\partial M}{\partial T}\right)^e=\frac{Y^3k^2\frac{\partial B_J(X)}{\partial X}\frac{B_J(X)}{T}\frac{\partial \eta}{\partial T}+Y^2k^2B_J(X)\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\frac{\partial \eta}{\partial H}}{1-Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}} \tag{B3}$$

it follows from (B2)

$$\left(1-Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}\right)\frac{\partial M}{\partial T}=\left(1-Y^2k\frac{\partial B_J(X)}{\partial X}\frac{\eta}{T}\right)\left(-Yk\frac{\partial B_J(X)}{\partial X}\frac{X}{T}\right)^c+Y^3k^2\frac{\partial B_J(X)}{\partial X}\frac{B_J(X)}{T}\frac{\partial \eta}{\partial T}+Y^2k^2B_J(X)\frac{X}{T}\frac{\partial B_J(X)}{\partial X}\frac{\partial \eta}{\partial H}.$$

This is identical to the right-hand side of Eq. (B1).

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